Creating Massive Entanglement of Bose-Einstein Condensed Atoms

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We propose a direct, coherent coupling scheme that can create massively entangled states of Bose-Einstein condensed atoms. Our idea is based on an effective interaction between two atoms from coherent Raman processes through a (two atom) molecular intermediate state. We compare our scheme with other recent proposals for the generation of massive entanglement of Bose condensed atoms.

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Entanglement lies at the heart of the difference between the quantum and classical multiparticle world. It is a phenomenon which facilitates quantum information and quantum computing with many qubits. Recently, several interesting developments have occurred in studies of massively entangled atomic states. Based on the proposals of Mølmer and Sørensen [1], a controlled, entangled state of 4-ions was successfully created by the NIST ion trap group [2]. Zeilinger and co-workers prepared three entangled photon or Greenberger-Horne-Zeilinger (GHZ) states by selecting from two beams of entangled photon pairs [3]. Entanglement between two atoms and a microwave photon was also detected in a "step-by-step" process [4].

Of these and other related developments, the idea of Mølmer and Sørensen [1] is especially interesting. They proposed a direct coupling to the multiparticle, entangled final state through a virtual, intermediate state, which was a common (quantum) mode of the motion of all the ions. Similar type interactions were also proposed by Milburn [5]. Both proposals allow for the creation of massive entangled states by unitary evolution, starting from certain pure initial states.

A Bose-Einstein condensate of a dilute atomic vapor is a convenient source of atoms, well approximated as initially being in pure and separable states. Sørensen *et al.* [6] suggested creating massively entangled, spin squeezed states from a two component condensate using the inherent atom-atom interactions. Spin-exchange collisional interactions in a spinor condensate were also proposed as a candidate for creating entangled pairs of atoms [7,8]. Most of these proposals work in the two mode approximation where one motional state is assumed for each spinor component of condensed atoms.

Raman transitions mediated by long-range dipole-dipole interactions have been proposed in entanglement schemes for quantum computing [9,10]. In this Letter, we propose a new type of coupling also based on the two atom, effective interaction from a Raman process through intermediate, molecular states. We show that our coupling applied to condensate atoms can achieve both massive entanglement, similar to that of Mølmer and Sørensen [1] and improved spin squeezing [6–8,11].

Consider a system (see Fig. 1) involving two, Λ-type atoms whose initial and final states are described by the same noninteracting atomic states $|g\rangle$ and $|g'\rangle$. Transitions between $|g\rangle$ and $|g'\rangle$ for the pair of atoms are coupled by two laser fields ($i = 1, 2$, frequency ω_i, wave vector $\vec{k}_i, k_i = \omega_i/c$, and Rabi frequency Ω_i) through an intermediate excited state, which is chosen to be a bound, molecular excited state.

We consider an excited molecular state $|e_b\rangle$ asymptotically connected, for large internuclear separation, to one ground $|\mu\rangle, \mu = g, g'$ and one excited atom $|e\rangle$. An example is the 0_g^- state, which has been extensively discussed in the context of photoassociation of ultracold atoms [12,13]. The recent experiment by Heinzen and co-workers on the production of ground state molecules from an atomic condensate by a two photon Raman

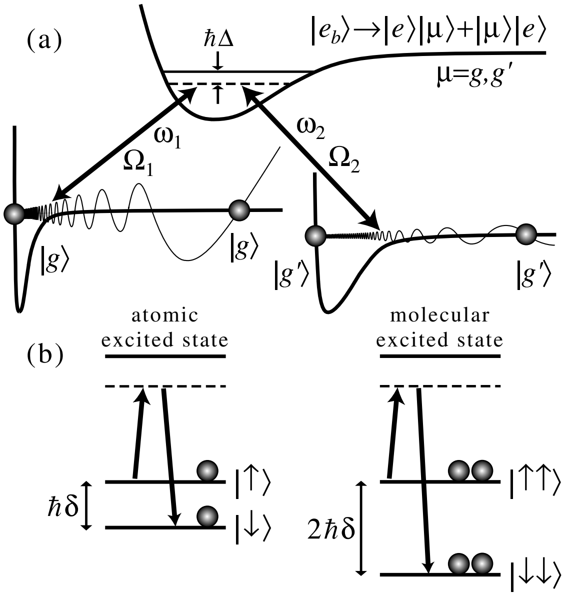

FIG. 1. (a) The general two photon Raman scheme via an intermediate, excited, molecular transition. (b) Raman coupling between two spin states is illustrated for the atomic and molecular cases. In the atomic case, single atoms can be prepared in a coherent superposition of spin up and spin down. In the molecular case, pairs of atoms are prepared in a coherent superposition of spin up and spin down.

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process [14] provides additional motivation to explore our ideas experimentally. The photoassociation process [12,13,15] used in Heinzen's experiment relies on the transition strength of going from a ground "free" (two atom) to a ground "bound" (molecular) state via an intermediate, excited bound (molecular) state. On the other hand, what we desire is a transition from a ground free to another ground free state via an intermediate, excited bound state.

The question of whether this is possible or not for a trapped ultracold cloud of atoms does not seem to depend on the sample density (in the weakly interacting limit), but will depend, to a large degree, on the trap strength and the excited, bound state structure. That is, we can determine the strength of the transition by considering the collision of a pair of atoms in the combined trap and molecular potentials and then summing over the number of pairs of atoms available.

Assuming all trapping potentials for the ground and excited states to be harmonic, there is a separation of the trapping potentials in terms of the center of mass $\vec{R}$ and relative coordinates $\vec{r}$ of the two atoms. When short-range interactions are approximated by their optical contact forms, Wilkens and co-workers provide analytic solutions for the center of mass and relative motion of two interacting atoms inside a harmonic trap [16]. In principle we can find all bound states in the $\vec{r}$ potential due to the external trapping potential, even with a realistic molecular interaction potential [17]. In general one can include all relative motional states and still be able to find the effective coupling between the selected electronic states [18]. We consider, however, only the lowest unbound state of the relative motion $|0\rangle$ in the electronic ground state, a situation well approximated by Bose condensed atoms.

If we choose a state fairly deep in the excited state molecular potential, then adjacent molecular states will be well separated in energy and we can consider coupling to only one intermediate bound state with vibrational quantum number m_b. Furthermore, for a sufficiently large detuning of the coupling lasers from the excited state $|m_b\rangle$, we can perform an adiabatic elimination of the excited state. Neglecting configurations not directly involved in the two photon process [19], the Hamiltonian for the two atoms initially in the g state can be written as

$$\mathcal{H} = \sum_{\mu=g,g'} \left[\frac{\vec{P}^2}{2(2M)} + V_{tR}(\vec{R}) + 2\hbar\omega_{\mu g} + E_0 \right] \times |\mu,\mu;0\rangle\langle\mu,\mu;0| + V_R(\vec{R},t), \qquad (1)$$

where

$$V_R = \frac{\hbar\Omega_R}{2}|\eta_{0m_b}|^2 e^{i\Delta\omega t} e^{i\vec{K}\cdot\vec{R}} |g',g';0\rangle\langle g,g;0| + \text{H.c.} \qquad (2)$$

$\Delta\omega = \omega_2 - \omega_1$, $\vec{K} = \vec{k}_1 - \vec{k}_2$, V_{tR} is the trapping potential for the center-of-mass motion, $\hbar\omega_{\mu g}$ is the energy difference between the atomic states g and g or g' and, for simplicity, we have assumed the same relative motional state $|0\rangle$, with energy E_0, for the pair of atoms in g or g' (i.e., atoms in state g or g' see the same trapping potential). Since $|e_b\rangle$ is asymptotically connected with $|e\rangle|\mu\rangle + |\mu\rangle|e\rangle$ (for $\mu = g, g'$) we can set the dipole matrix element $\vec{d}_i = \vec{d}_i' = \vec{d}$, for the $|\mu\rangle$ to $|e\rangle$ transition of atom i. Then $\Omega_R = \Omega_1\Omega_2^*/2\Delta$ is simply the two photon Rabi frequency coupling between atomic states g and g'. Typically $|\vec{k}_1| \approx |\vec{k}_2|$ and we can use $\eta_{0m_b} = \langle 0|\cos(\vec{k}_1 \cdot \vec{r})|m_b\rangle$ for both free-bound transition amplitudes. $|\eta_{0m_b}|^2$ is essentially the Franck-Condon factor, or a measure of the strength of the free-bound transition and will vary considerably with $\vec{r}$ for small values of r, due to the relatively short range of the molecular interactions. It is therefore important to pick the intermediate state $|m_b\rangle$ which results in a large $|\eta_{0m_b}|$. These values can be determined for selected molecular states from the results of photoassociation experiments. They can also be computed directly if accurate potentials are available. Detailed discussions are given in [13,15].

For two counterpropagating waves ($\vec{K}/2 \approx \vec{k}_1 \approx -\vec{k}_2$) and $\mu = g$, the above result is similar to the single atom case often referred to as Bragg diffraction [20]. The elementary process involves the simultaneous absorption and stimulated emission of two photons. But, in contrast to the single atom case, a pair of atoms are now involved. Hence for Bose condensed atoms with $p_i \approx 0$, this coupling produces pairs of atoms in the state $\alpha|p_1 \approx 0, p_2 \approx 0\rangle + \beta|\vec{p}_1 = \hbar\vec{K}/2, \vec{p}_2 = \hbar\vec{K}/2\rangle$. The momentum shift per atom and the resonance (energy conservation) condition $\Delta\omega$ are at half the values for atomic Bragg diffraction.

For a Raman process with two copropagating waves ($\vec{k}_1 \approx \vec{k}_2, \vec{K}/2 \approx 0$) between two nearly degenerate states g and g' [as depicted in Fig. 1(b)], the resonance condition is $\Delta\omega = 2\omega_{g'g}$, twice the atomic resonance. Note that, in this case, there is no $\vec{R}$ dependence in V_R.

We now consider many atoms in a spin 1/2 system to investigate the extent to which Raman coupling via an intermediate molecular state can produce spin squeezing [11] and correspondingly massive entanglement [6]. If we designate $|g\rangle$ and $|g'\rangle$ as $|\uparrow\rangle$ and $|\downarrow\rangle$ for spin up and down, respectively, our Raman coupling for the two particle case is of the form

$$\frac{\hbar\Omega_R}{2}[(|\uparrow\rangle\langle\downarrow|)_1 \otimes (|\uparrow\rangle\langle\downarrow|)_2 + (|\downarrow\rangle\langle\uparrow|)_1 \otimes (|\downarrow\rangle\langle\uparrow|)_2] = \frac{\hbar\Omega_R}{2}(\sigma_x^{(1)} \otimes \sigma_x^{(2)} - \sigma_y^{(1)} \otimes \sigma_y^{(2)}), \qquad (3)$$

where $\sigma_{\mu=x,y,z}$ are the Pauli matrices.

We first distinguish our coupling scheme from other relevant models. In the original scheme of Mølmer and Sørensen [1], the two atom coupling takes the form $\frac{1}{2}\hbar\Omega_R\sigma_x^{(1)} \otimes \sigma_x^{(2)}$, a form different from ours. If the pairwise interaction acts indiscriminately for all pairs of atoms, it is convenient to analyze the effect of such couplings for many atoms in terms of a collective spin operator $J_\mu = \sum_i \sigma_\mu^{(i)}$, $\mu = x, y, z$, where the sum is over the number of atoms N [21,22]. One can then show that the Hamiltonian for the Mølmer and Sørensen scheme becomes $V_M = \sum_{i<j} \frac{1}{2}\hbar\Omega_R \sigma_x^{(i)} \otimes \sigma_x^{(j)} = \frac{1}{4}\hbar\Omega_R (J_x^2 - N)$. In contrast, our scheme gives

$$\sum_{i<j} \frac{\hbar\Omega_R}{2} (\sigma_+^{(i)} \otimes \sigma_+^{(j)} + \sigma_-^{(i)} \otimes \sigma_-^{(j)}) = \frac{\hbar\Omega_R}{2} (J_x^2 - J_y^2). \tag{4}$$

Recently, a many body, two mode coupling scheme was proposed by Sørensen *et al.* [6]. They considered a two component (i.e., $|\downarrow\rangle$ and $|\uparrow\rangle$) condensate weakly interacting via s-wave collisions described by a mean field. In the approximation where each component has the same spatial mode, the interaction in terms of the collective spin operators $J_{x,y,z}$ is of the form $V_S = \frac{1}{2}\hbar\Omega_R J_z^2$.

In order to compare the various coupling schemes, we investigate the time evolution assuming a pure initial state with a fixed, total number of atoms N. Using the notation of second quantization, where $a_j^\dagger$ and a_j are the operators for creating and annihilating a particle in state j ($j = \uparrow, \downarrow$), the collective spin operators can be written as $J_x = (a_\downarrow^\dagger a_\uparrow + a_\uparrow^\dagger a_\downarrow)/2$, $J_y = i(a_\downarrow^\dagger a_\uparrow - a_\uparrow^\dagger a_\downarrow)/2$, and $J_z = (a_\uparrow^\dagger a_\uparrow - a_\downarrow^\dagger a_\downarrow)/2$. To numerically calculate the time evolution, we expand the wave function as $|\psi\rangle = \sum_{m=0}^{N} c_m(t)\,|m\rangle_\downarrow |N - m\rangle_\uparrow$, where $|m\rangle_j = (a_j^\dagger)^m |\text{vac}\rangle / \sqrt{m!}$, with the initial conditions given by the $c_m(0)$s.

For N an even integer [23], the time evolution operator for V_M takes a simple, analytical form at $\Omega_R t = \pi/2$, producing a massive GHZ-type wave function $|\text{GHZ}\rangle_N = [|N\rangle_\uparrow |0\rangle_\downarrow + \eta |0\rangle_\uparrow |N\rangle_\downarrow]/\sqrt{2}$, where η is purely a phase factor. While the Mølmer and Sørensen coupling (V_M) produces perfect GHZ-type states at selected times, the Sørensen *et al.* spin squeezing scheme (V_S) and our scheme [Eq. (4)], in general, do not produce exact GHZ-type states. From numerical simulations, however, we find that our model can produce more than 50% overlap with the state $|\text{GHZ}\rangle_N$ at selected times (see Fig. 2).

We can also compare the achievable spin squeezing between our scheme [Eq. (4)] and that of Sørensen *et al.* (V_S), using the squeezing parameter $\xi^2 = N(\Delta J_{\vec{n}_1})^2 / (\langle J_{\vec{n}_2}\rangle^2 + \langle J_{\vec{n}_3}\rangle^2)$, where $\vec{n}_i, i = 1, 2, 3$ are mutually orthogonal unit vectors [6,11].

Sørensen *et al.* [6] have shown that $\xi^2(t > 0) < 1$ for some set of $\vec{n}_i$'s. Their scheme (V_S) is in fact the one-axis twisting model considered by Kitagawa and Ueda earlier [11]. In this case the problem can be solved analytically, with the result $(\Delta J_{\vec{n}_1}^2)_{\min} \sim N^{1/3}$.

On the other hand, our coupling resembles the two-axis countertwisting model of Kitagawa and Ueda [11], and has to be solved numerically. In the limit of large N and with the condensate initially in one spin state, $J_z = -N/2$, one can show that $(\Delta J_{\vec{n}_1}^2)_{\min} \approx 1/2$. The optimal squeezing in this case occurs along $\hat{x} + \hat{y}$. This result can be easily verified by making a semiclassical approximation in the dynamical equations for J_x and J_y. We find that the time scale of reaching maximum squeezing is $\sim 1/(N\Omega_R)$ (see also [24]). For condensates containing 10^6 atoms, even with a very weak coupling $\Omega_R \approx 1$ (Hz), the maximum squeezing is reached within a microsecond.

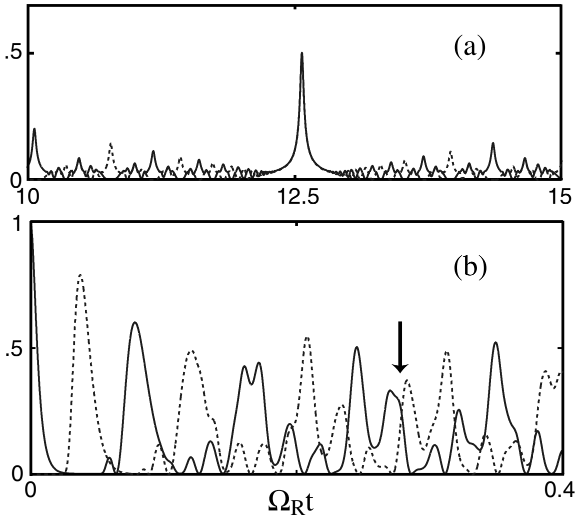

FIG. 2. The time dependent probabilities $|c_0|^2$ (solid line) and $|c_N|^2$ (dashed line) of being in states $|N\rangle_\downarrow |0\rangle_\uparrow$ and $|0\rangle_\downarrow |N\rangle_\uparrow$, respectively, for (a) the Mølmer and Sørenson coupling (V_M) and (b) our "molecular Raman" coupling [Eq. (4)]. The initial conditions are $c_m(0) = \delta_{0m}$. The number of atoms in the simulation is $N = 10^3$. The projection onto the $|\text{GHZ}\rangle_N$ state corresponds to $|c_0|^2 = |c_N|^2$. In (a) it is apparent that the coupling V_M produces a perfect $|\text{GHZ}\rangle_N$ state at $\Omega_R t = 4\pi$. While in (b) it appears that at $\Omega_R t \approx 0.28$ (indicted by the arrow) our coupling scheme results in about 50% of the atoms in the $|\text{GHZ}\rangle_N$ state.

In Fig. 3, we show the numerically computed, minimum squeezing parameters, ξ^2, as a function of time for our coupling scheme [Eq. (4)] and for the Sørensen *et al.* scheme (V_S). In contrast to the Sørensen *et al.* scheme, our scheme achieves better squeezing at an earlier time. In addition for V_S, the direction $\vec{n}_1$ along which minimum squeezing is observed varies with time [6,11]. While for the coupling of Eq. (4), it is fixed along $\hat{x} + \hat{y}$ [11].

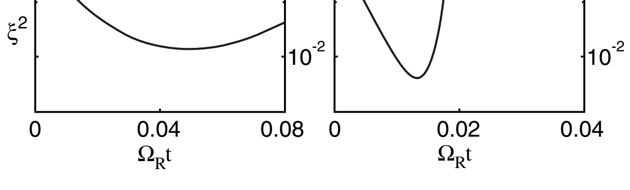

FIG. 3. The time evolution of the minimum spin squeezing parameter ξ^2 for (a) the coupling V_S and (b) the coupling given by Eq. (4).

In order to realize our coupling scheme experimentally, the molecular coupling [Fig. 1(b), right side] needs to dominate over the atomic coupling [Fig. 1(b), left side]. That is, we need to achieve a molecular coupling $\Omega_R^M = \Omega_1\Omega_2^*|\eta_{0m_b}|^2/\Delta_M \gg \Omega_R^A = \Omega_1\Omega_2^*/\Delta_A$, where $\Delta_{M/A}$ are detunings from the molecular and atomic intermediate states, respectively. We should also have $\Delta_M \gg \gamma_M$, the excited molecular state linewidth, to minimize spontaneous emission, which would lead to decoherence and loss of atoms. These constraints suggest that a deep molecular bound state with significant transition strength should be chosen in order to maximize the detuning from the atomic transition and achieve a sufficient detuning from excited molecular states. In addition, if $\Delta_A \gg \Delta_M$, further suppression of the atomic transition might be possible by a suitable choice of laser polarizations.

Another mechanism for suppressing the atomic coupling with respect to the molecular coupling is the two photon, Raman resonance detuning. For Bragg diffraction, the transition via a molecular coupling occurs at half the detuning of the transition via an atomic coupling (50 vs 100 kHz, respectively for sodium). If the Raman transition involves changes in the internal state of the atom, then the frequency for the Raman transition via a molecular coupling will be at twice the frequency of the Raman transition via an atomic coupling [see Fig. 1(b)]. For Raman transitions between Zeeman sublevels in modest magnetic fields this frequency difference can be several MHz, which would greatly suppress the single atom transition.

There are several advantages of our coupling scheme. The different Raman resonance frequency is a clear signature for the Raman transition via a molecular intermediate state. In addition, for Bragg diffraction via the molecular coupling, the atoms would move at half the speed of atoms that have undergone atomic Bragg diffraction.

Our scheme is based on an engineered interaction that can be turned on and off similar to the scheme of Mølmer and Sørensen (V_M). In contrast to the Sørensen *et al.* coupling scheme (V_S), our scheme works for noninteracting ground state atoms, which can decrease the noise due to atom-atom interactions in a U(1) symmetry breaking condensate state. Our scheme also achieves the same level of squeezing and the same high value of overlap with the massive GHZ state, even as the number of atoms is increased.

Finally, we note that our discussions above can also be applied to a pair of a different species of atoms. For example, Raman coupling using a molecular intermediate state of the Li-Cs dimer could create entangled pairs of Li and Cs.

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